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(72) Inventor: MISPREUVE, Henri, L., S., A.; Chemin des Vergers 11, CH-1197 Prangins (CH).			
(74) Agent: DAMOCLES, Nemia, C.; The Dow Chemical Company, Patent Dept., P.O. Box 1967, Midland, MI 48641-1967 (US).			
(54) Title: REDUCED DENSITY FLEXIBLE POLYURETHANE FOAM FROM METHYLENE DIPHENYLISOCYANATE			
(57) Abstract			
<p>Disclosed is the preparation of a water-blown, methylene diphenylisocyanate-based (MDI) polyurethane foam, and particularly a flexible, slabstock polyurethane foam. The foam is prepared in the presence of a processing aid which comprises at least one monohydroxyl polyether having an equivalent weight of from 600 to 5000 and an oxyethylene content of from 25 to 75 weight percent. The processing aid permits the more efficient use of water as blowing means providing a foam having a reduced density and attractive physical properties.</p>			

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REDUCED DENSITY FLEXIBLE POLYURETHANE FOAM FROM METHYLENE
DIPHENYLISOCYANATE

This invention relates to the preparation of a water-blown, methylene
5 diphenylisocyanate-based (MDI) polyurethane foam, and particularly a flexible, slabstock
polyurethane foam.

Polyurethane foam manufacturers frequently desire, for economic reasons, to
prepare foam having a lower density without detriment to competitive physical properties. An
obvious means of attaining a lower density foam is to increase the amount of blowing agent
10 present during the manufacturing process when reacting a polyisocyanate with a polyether or
polyester polyol. Traditionally volatile organic compounds such as, for example,
trichlorofluoromethane or methylene chloride have been employed as "physical" blowing
agents when preparing flexible polyurethane foam. However, due to a growing concern for
the well being of the environment, the use of many physical blowing agents has largely ceased
15 with only water being used in preference as a "chemical" blowing means. Water reacts with
the polyisocyanate generating *in situ* carbon dioxide which then functions as the blowing
agent.

Previously, simply increasing the amount of physical blowing agent present
generally provided a direct reduction of density of the resulting foam. However, it has been
20 found that the anticipated foam density reduction is frequently not attained when water is
used as the blowing agent. This is particularly the case when desiring to prepare foam having a
density of from 20 to 60 kg/m³ and when such foam is prepared from methylene
diphenylisocyanate instead of toluene diisocyanate. Furthermore, physical foam properties
frequently become inferior and are not so commercially attractive. When preparing flexible
25 slabstock polyurethane foam, it is considerably more convenient to use methylene
diphenylisocyanate instead of toluene diisocyanate. Use of the latter isocyanate requires
taking extensive precautions to minimize and avoid exposure of persons to toluene
diisocyanate emissions. Accordingly it would be desirable to provide a flexible slabstock
polyurethane foaming process that permits the manufacture of a low density methylene
30 diphenylisocyanate-based polyurethane foam in the presence of water.

As polyisocyanates are generally expensive, there is a strong commercial interest
to reduce the required amounts when preparing the foam. However at reduced
polyisocyanate amounts, particularly at an isocyanate reaction index of less than 100, there is
often an unacceptable loss of physical properties. Accordingly, it would also be desirable if
35 such process could also permit the preparation of the polyurethane foam at a lower isocyanate
reaction index without detriment to the physical properties of the resulting foam. For this
purpose, the preparation of water-blown flexible polyurethane foam from methylen

diphenylisocyanate at a reduced reaction index and in the presence of a processing aid comprising a monohydroxyl polyether has been investigated.

The use of a monohydroxyl polyether when preparing flexible polyurethane foam is reported in the open literature. See, for example, U.S. Patent 3,405,077 which discloses the use of monoalcohols having a hydroxyl equivalent weight of from 74 to 150 in combination with polyether polyols for the preparation of soft polyurethane foams. Similarly, Canadian Patent 787,977 teaches the use of low molecular weight monoalcohols for preparing soft polyurethane foam in the presence of mainly trichlorofluoromethane as the blowing agent. Use of monohydroxyl polyethers when preparing flexible polyurethane foam is also disclosed in U.S. Patents 4,981,880; 4,950,694; and 4,950,695.

The preparation of flexible slabstock polyurethane foam with polyol compositions containing monohydroxyl polyoxypropylene ethers of equivalent weights up to 719 is taught by Arceneaux et al, Journal of Elastomers and Plastics, p. 63, Vol. 14 (1982). The prepared flexible slabstock polyurethane foam has improved elongation properties but unacceptable load bearing, tear resistance, and compressive and tensile strength properties.

It has been found that by using a combination of a selected monohydroxyl polyether processing aid with a selected polyol in the preparation of a polyurethane foam that a more efficient use of water as blowing means is obtained. This finding permits the manufacture of flexible polyurethane foam having a reduced density while retaining attractive physical properties.

In one aspect, this invention relates to a process for preparing a flexible polyurethane foam which comprises reacting a polyisocyanate with a polyol in the presence of a processing aid and water, wherein:

- a) the polyisocyanate comprises a methylene diphenylisocyanate, or a modified methylene diphenylisocyanate having an isocyanate content of at least 18 weight percent;
- b) the polyol comprises at least one polyoxypropylene-oxyethylene polyol containing at least two hydroxyl groups, said polyol having an average equivalent weight of from 1600 to 5000, an average oxyethylene content of at least 10 weight percent based on its total weight, and an average primary hydroxyl content of at least 55 percent of its total hydroxyl content; and
- c) the processing aid comprises at least one monohydroxyl polyether having an equivalent weight of from 600 to 5000 and an oxyethylene content of from 25 to 75 weight percent, and

wherein the processing aid is present in an amount of from 1 to 20 parts per 100 parts by weight of polyol; the water is present in an amount of from 2 to 10 parts per 100 parts by

weight of polyahl; and the polyisocyanate is present in an amount sufficient to provide an overall isocyanate reaction index from 50 to 90.

In another aspect, this invention relates to a flexible polyurethane foam obtained according to the above-defined process.

5 In yet another aspect, this invention relates to an isocyanate-reactive composition suitable for preparing a flexible polyurethane foam which contains a polyahl, a processing aid, and water wherein:

- 10 (a) the polyahl, comprising a polyoxyalkylene polyol, has at least two isocyanate-reactive hydrogen atoms/molecule, an average equivalent weight of from 1600 to 5000, an average oxyethylene content of at least 10 weight percent based on its total weight, and an average primary hydroxyl content of at least 55 percent of its total hydroxyl content;
- (b) the processing aid, comprising one or more monohydroxyl polyethers having an equivalent weight of from 600 to 5000 and an oxyethylene content of from 25 to 15 75 weight percent, is present in an amount of from 1 to 20 parts per 100 parts by weight of (a); and
- (c) the water is present in an amount of from 2 to 10 per 100 parts by weight of (a).

This invention broadly relates to a flexible methylene diphenylisocyanate (MDI)-based polyurethane foam, preferably all water blown, and especially a flexible slabstock 20 polyurethane foam exhibiting attractive load bearing properties and a reduced density. By "reduced density," it is understood that the foam has a lower density in comparison to foam obtained from conventional art processes using the same amounts of blowing agents providing the same blowing potential. The free rise density of such foam typically is from 15 to 60 kg/m³, and preferably from 20 to 45 kg/m³. The load bearing properties of the foam may be 25 characterized by a modulus or "SAG" factor performance generally of 2.5 or higher, and frequently of 3.0 or higher.

Generally, such polyurethane foam can be prepared by reacting a polyisocyanate with a polyahl in the presence of a processing aid and water, and optionally catalyst, surfactant, flame retardant and other additives well known to those skilled in the art of 30 preparing polyurethane foam. The components employed in such a process are described more fully hereinafter.

The Polyisocyanate

The polyisocyanate used in the present invention comprises a methylene diphenylisocyanate (MDI) or a urethane-modified adduct thereof having an isocyanate content 35 of at least 18 weight percent.

MDI includes the isomers 4,4'- and 2,4'-methylene diphenylisocyanate in a weight ratio of advantageously from 98:2 to 50:50, preferably from 95:5 to 70:30, and more preferably from 90:10 to 70:30. Such a ratio is

desirable to provide optimum physical properties including the SAG factor of the resulting foam. While it is preferred to use a polyisocyanate that consists essentially of 4,4'- and 2,4'-MDI isomers, such MDI isomers may also be used in admixture with polymeric polymethylene polyphenylisocyanate. When present, the amount of polymeric polymethylene polyphenylisocyanate in the total polyisocyanate mixture will be less than 30, and preferably less than 15 percent based on parts by weight of total polyisocyanate present.

The urethane-modified adduct of MDI is obtained by reacting MDI with a polyoxyalkylene diol or triol, present in a substoichiometric amount, to provide the resulting adduct with an isocyanate content advantageously of from 20, preferably from 25, more preferably from 28, and preferably to 31, more preferably to 30 weight percent. Suitable and preferred urethane-modified polyisocyanates are disclosed in U.S. Patent 5,114,989 and exemplified by, for example, the urethane-modified polyisocyanate designated as ISONATE M100 available from The Dow Chemical Company.

In the process, the polyisocyanate is present in an amount sufficient to provide an isocyanate reaction index of typically from 50 to 90, preferably from 65, more preferably from 70, and preferably up to 85. An isocyanate reaction index of 100 corresponds to one isocyanate group per isocyanate reactive hydrogen atom including those present from the polyahl, processing aid and water.

The Polyahl

The polyahl comprises at least one polyoxypropylene-oxyethylene polyol containing two or more hydroxyl groups, and preferably comprises at least two different polyoxypropylene-oxyethylene polyols. The polyahl can additionally include a variety of compounds and substances containing isocyanate-reactive hydrogen atoms such as amines, thiols, carboxylic acids and polyester polyols.

The polyahl is characterized by its average equivalent weight, average oxyethylene content and average primary hydroxyl content. In general, the polyahl has an average equivalent weight of from 1600 to 5000, preferably from 1650, more preferably from 1700, preferably to 4000, more preferably to 3000. The polyahl has an average oxyethylene content of at least 10, preferably from 12, and more preferably from 14, preferably to 50, more preferably to 40, and most preferably to 30 percent based on its total weight. Although a higher oxyethylene content may be contemplated, the processing and resulting foam properties may not be commercially attractive. The oxyethylene content of the polyahl is understood to provide a good compatibility with water as present in the process of the invention. The polyahl has an average primary hydroxyl content of at least 55, preferably at least 60, more preferably at least 65 percent; preferably to 90, and more preferably to 85 percent of its total hydroxyl content.

In addition to the above features, the polyahl advantageously has an average functionality of at least 2, preferably from 2.25 to 6, and more preferably from 2.25 to 4, and

yet more preferably to 3.5. The term "functionality" is intended to mean the average number of isocyanate-reactive hydrogen atoms/molecule.

As mentioned, the polyahl comprises at least one polyoxypropylene-oxyethylene polyol. Such polyol can be defined by the above-mentioned equivalent weight, oxyethylene content and primary hydroxyl percentage.

In a preferred embodiment of this invention, the polyahl comprises a mixture of (i) a first polyoxyalkylene-oxyethylene polyol and (ii) a second polyoxyalkylene-oxyethylene polyol. Such a mixture advantageously, based on total weight of polyol (i) and polyol (ii) present, contains polyol (i) in an amount of from 60 to 98, preferably from 70 to 98, more preferably from 80 to 98, and most preferably from 80 to 95 weight percent; and polyol (ii) in an amount of from 2 to 40, preferably from 2 to 30, more preferably from 2 to 20, and most preferably from 5 to 15 weight percent. In such a mixture, polyol (i) is as described above but limited to an oxyethylene content of less than 50, preferably from 10 to 40 weight percent; and advantageously has a functionality of from 2 to 6, preferably from 2 to 4. Polyol (ii) is distinguished from polyol (i) in that it has an oxyethylene content of more than 50, preferably at least 60, and preferably up to 100, more preferably up to 85 weight percent; and an equivalent weight of from 500 to 5000, preferably from 1000 to 3000. Polyol (ii) advantageously has a functionality of from 2 to 6, preferably from 2 to 4.

Suitable polyoxyalkylene polyols which may constitute the polyahl include those which can be obtained by reacting an initiator containing two or more active hydrogen atoms with alkylene oxides comprising ethylene oxide, preferably in combination with propylene oxide or butylene oxide. Typical initiators include water, alkylene glycols, glycerine, trimethylolpropane, pentaerythritol, methylglucoside, sorbitol, mannitol, and sucrose. In addition to these initiators, other suitable initiators include organic amines such as ethylene diamine and toluene diamine; alkanolamines such as mono- or diethanolamine or diisopropanolamine; and aromatic resins such as phenol/formaldehyde condensates and phenol/formaldehyde/alkanolamine condensates. Preferred initiators include water, alkylene glycols, glycerine, trimethylolpropane and pentaerythritol. The preparation procedures for the polyols suitable for use in the process of the invention are well known to those skilled in the art and do not require further explanation. Exemplary of commercially available polyether polyols corresponding to the above-defined components include, for example, for polyol (i), products designated as VORANOL EP1900, VORANOL CP6008 and VORALUX HP700; and for polyol (ii), VORANOL CP1421, all sold by The Dow Chemical Company.

When desiring to enhance the load bearing properties of the obtained foam, advantageously the polyahl may further contain a stably dispersed particulate polymer in an amount of from 2 to 20, preferably from 4 to 12 weight percent, based on the total weight of the polyahl including particulate polymer. Suitable particulate polymers include polyurea (PHD), polyisocyanate - polyamine adducts (PIPA) and particularly styrene/acrylonitrile (SAN)

adducts. Polyoxyalkylene polyols containing the particulate polymer can be prepared by, for example, *in situ* polymerizing of styrene with acrylonitrile in a polyoxyalkylene polyol corresponding, for example, to the above-described polyol (i) or (ii). Exemplary of commercially available polyether polyols further comprising a stably dispersed particulate polymer and suitable for use as a polyahl component in the present invention include for example products designated as VORALUX HN 201 available from The Dow Chemical Company.

The Processing Aid

The present invention requires the presence of a processing aid to assist in the manufacture of the foam and additionally to enhance the SAG performance of the resulting foam. The processing aid is present in an amount of from 1 to 20, preferably from 5, more preferably from 7, and preferably up to 15, and more preferably up to 12.5 parts per 100 parts by total weight of the polyahl. The processing aid comprises one or more monohydroxyl polyether substances having an equivalent weight of from 600 to 5000, preferably from 1000 to 4000, and more preferably from 1500 to 3500. The monohydroxyl polyether has an oxyethylene content of from 25 to 75, preferably from 30 to 60, and more preferably from 35 to 55 percent based on its total weight, the balance comprising oxypropylene or oxybutylene units. Commercially available substances suitable for use as processing aids of this invention include products sold by The Dow Chemical Company under the trademark of SYNALOX such as, for example, SYNALOX 25-50B and especially SYNALOX 25-300B.

The Blowing Agent

The blowing agent used in the present invention comprises water which reacts with polyisocyanate leading to the *in situ* production of carbon dioxide. The water is present in an amount of from 2 to 10, preferably from 2.5 to 8, more preferably to 6.5, and most preferably to 5.5 parts per 100 parts by weight of total polyahl. In a highly preferred embodiment of this invention, the blowing agent consists of water. In a lesser preferred embodiment, the blowing capacity provided by water may be supplemented by physical blowing agents. Exemplary of such physical blowing agents include fluorocarbons and chlorofluorocarbons such as, for example, dichlorotrifluoroethane (R-123), dichlorofluoroethane (R-141a), chlorodifluoroethane (R-142b), tetrafluoroethane (R-134a), and chlorodifluoromethane (R-22); hydrocarbons such as butane, pentane, cyclopentane, hexane and cyclohexane; and entrained gases such as air, argon and nitrogen. Typically, the blowing agent is present in an amount to confer to the resulting foam a free rise density as already mentioned.

Further to the above-mentioned constituents, optionally present in the foaming process are other substances including chain extending agents, crosslinking agents, urethane promoting catalysts, foam stabilizing agents and flame retardants. Typical of chain extending agents and crosslinking agents include, for example, glycerine and diethanolamine. Foam stabilizing agents include silicon surfactants, for example, siloxane-oxyalkylene copolymers

- such as products sold under the trademark TEGOSTAB by Th. Goldschmidt AG including B-4113, B-4690 and B-8681; and products sold by Air Products including the product designated as DABCO DC 5258. Suitable catalysts which may be used to promote the formation of urethane groups include tertiary amines and organometallic compounds especially tin compounds.
- 5 Exemplary of tertiary amine compounds include N,N-dimethylcyclohexylamine, N,N-dimethylbenzylamine, N,N-dimethylethanolamine, bis(dimethylaminoethyl)ether and 1, 4-diazobicyclo[2,2,2]octane; of tin compounds include stannous octoate and dibutyltin dilaurate. Combinations of amine and/or tin compounds as catalyst may advantageously be present. When it is desired to impart a degree of flame retardancy to the polyurethane foam
- 10 present can be antimony-, phosphorus- or nitrogen-containing substances including for example, melamine, tris(chloroethyl)phosphonate or preferably halogen-free phosphorus compounds including for example triethylphosphate.

The following examples are illustrative of the present invention. Unless otherwise stated, all parts and percentages are by weight. Where reported, properties of

15 foams as obtained are observed according to the following published test procedures; tensile strength and elongation - DIN 53571; compression load deflection (CLD)- DIN 53577; indentation load deflection (ILD)- DIN 53576; resilience - ASTM 3574-86. The components of the formulations are as identified hereinafter:

- MDI - A: ISONATE M100, a proprietary urethane-modified MDI composition
- 20 with an NCO content of 29 weight percent, available from The Dow Chemical Company.
- Polyol 1: A glycerine-initiated polyoxypropylene-oxyethylene polyol of 2000 equivalent weight, with an oxyethylene content of 14.5 percent, and primary OH content of 80 percent.
- 25 Polyol 2: A glycerine-initiated polyoxypropylene-oxyethylene polyol of 1600 equivalent weight, with an oxyethylene content of 17 percent, and primary OH content of 85 percent, and further containing 21 weight percent of a particulate styrene/acrylonitrile polymer
- Polyol 3: A glycerine-initiated polyoxypropylene-oxyethylene polyol of 1670
- 30 equivalent weight with an oxyethylene content of 75 percent, and primary OH content of 50 percent.
- Processing
- Aid - 1 : SYNALOX 25-3008 a proprietary monohydroxyl polyether, available from The Dow Chemical Company, and understood to have a molecular weight
- 35 of 3000 and an oxyethylene content of 50 percent.
- Catalyst 1: DABCO 33LV, an amine-based proprietary urethane-promoting catalyst available from Air Products.

Catalyst 2: NIAX A-1, an amine-based proprietary urethane-promoting catalyst available from OSI Specialities.

Catalyst 3: Stannous Octoate.

Surfactant 1: TEGOSTAB B 8681, a silicone-based proprietary surfactant available from Th Goldschmidt AG.

Surfactant 2: DABCO DC 5258, a silicone-based proprietary surfactant available from Air Products.

Example 1

Flexible, slabstock, polyurethane foam is prepared, according to the formulation given in Table I using a high pressure dispenser unit operating at 2900 rpm, with all component streams at 20°C, and with a polyahl output of 25 kg/min. Properties of the resulting foam is reported in Table I.

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Table I

	Foam A*	Foam 1	Foam B*	Foam 2	Foam 3	Foam C*	Foam 4	Foam 5	Foam 6	Foam 7	Foam 8	Foam D*	Foam 9
MDI type	A	A	A	A	A	A	A	A	A	A	A	A	A
INDEX	85	85	85	85	85	70	70	70	80	65	65	110	85
Polyol 1	100	95	100	95	90	50	45	95	95	95	95	95	95
Polyol 2	/	/	/	/	/	50	45	/	/	/	/	/	/
Polyol 3	/	/	/	/	/	/	5	5	5	5	/	/	/
POLYAHL: average Equivalent wt.	2000	2000	2000	2000	2000	1800	1700	1983	1983	1983	2000	2000	2000
Oxyethylene wt%	14.5	14.5	14.5	14.5	14.5	15.8	18	17.5	17.5	17.5	14.5	14.5	14.5
Primary OH %	80	80	80	80	80	82.5	76.5	78.5	78.5	78.5	80	80	80
Processing Aid - 1	/	5	/	5	10	/	5	5	5	5	5	5	5
Water	2.5	2.5	3	3	3	4.25	4.25	4.25	4.25	6.5	6.5	3.5	3.5
Diethanolamine	1	1	2.5	2.5	2.5	4	4	4	4	5	5	3	3
Glycerine	/	/	/	/	/	/	/	/	/	0.5	0.5	/	/
Catalyst 1	0.12	0.12	/	/	/	0.15	0.15	0.15	0.15	0.12	0.12	0.15	0.15
Catalyst 2	0.04	0.04	0.1	0.1	0.1	0.05	0.05	0.05	0.05	0.04	0.04	0.05	0.05
Catalyst 3	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.2	0.2	0.2	0.2	0.2	0.2
Surfactant - 1	0.8	0.8	0.3	0.3	0.3	0.5	0.5	/	/	/	/	/	/
Surfactant - 2	/	/	/	/	/	/	/	1.5	1.5	1.5	1.5	1.5	1.5
Foam Density (kg/m ³)	59.2	55.7	41.4	39.1	36.3	40.9	36.7	29.7	33.7	24.5	23.9	38	37.8
Tensile Strength (kPa)	65	65	79	47	59	108	58	/	/	/	/	/	/
Elongation (%)	156	143	110	117	77	114	96	/	/	/	/	/	/
Tear Resistance (N/m)	150	180	103	98	85	345	191	/	/	/	/	/	/
Resilience (%)	56	54	61	57	51	52	49	48	53	52	50	55	53
Compression Set (75 %)	3.2	3.3	4.5	7.5	6.8	18.1	24	/	/	/	/	/	/
CLD (40%) kPa	3.1	2.8	1.8	1.4	1.6	3.1	1.5	0.9	1.08	0.8	0.88	2.46	1.8
ILD Modulus (65/25)	3	3.3	3.2	3.6	4	2.7	2.9	3.7	4.2	4.1	3.6	3.5	3.3

* Not an example of this invention

1. A process for preparing a flexible polyurethane foam which comprises reacting a polyisocyanate with a polyahl in the presence of a processing aid and water, wherein:

- 5 a) the polyisocyanate comprises methylene diphenylisocyanate or a modified methylene diphenylisocyanate having an isocyanate content of at least 18 weight percent;
- 10 b) the polyahl comprises at least one polyoxypropylene-oxyethylene polyol containing at least two hydroxyl groups, said polyahl having an average equivalent weight of from 1600 to 5000, an average oxyethylene content of at least 10 weight percent, based on its total weight, and an average primary hydroxyl content of at least 55 percent of its total hydroxyl content; and
- 15 c) the processing aid comprises at least one monohydroxyl polyether having an equivalent weight of from 600 to 5000 and an oxyethylene content of from 25 to 75 weight percent, and wherein the processing aid is present in an amount of from 1 to 20 parts per 100 parts by weight of polyahl; the water is present in an amount of from 2 to 10 parts per 100 parts by weight of polyahl; and the polyisocyanate is present in an amount sufficient to provide an overall isocyanate reaction index of from 50 to 90.

2. The process of Claim 1 wherein the polyahl has an average equivalent weight of from 1650 to 4000, and an average primary hydroxyl content of at least 60 percent.

3. The process of Claim 1 wherein the polyahl contains at least two different polyoxypropylene-oxyethylene polyols which based on total parts by weights of (i) and (ii) present, comprises:

- 25 i) from 60 to 98 weight percent of a polyoxypropylene-oxyethylene polyol having an equivalent weight of from 1650 to 5000 and an oxyethylene content of less than 50 weight percent; and
- ii) from 2 to 40 weight percent of a polyoxypropylene-oxyethylene polyol having an equivalent weight of from 500 to 5000 and an oxyethylene content of more than 50 weight percent.

30 4. The process of Claim 3 wherein (i) is a polyoxyalkylene polyol having an equivalent weight of from 1650 to 3000, a nominal functionality of from 2 to 4, and an oxyethylene content of from 10 to 40 weight percent.

5. The process of Claim 3 wherein (ii) is a polyoxyalkylene polyol having an equivalent weight of from 1000 to 3000, a nominal functionality of at least 2, and an oxyethylene content of at least 60 weight percent.

6. The process of Claims 1 or 3 wherein the polyahl further comprises a stably dispersed particulate polymer.

7. The process of Claims 1 or 3 wherein the polyisocyanat is a urethane-modified polyisocyanate having an average isocyanate content of from 20 to 29 weight percent and prepared from a mixture of 4,4'- and 2,4'-methylene diphenylisocyanate, present at a weight ratio of from 98:2 to 50:50.

5 8. A flexible polyurethane foam obtained according to the process of Claims 1 to 7.

9. An isocyanate-reactive composition suitable for preparing a flexible polyurethane foam which contains a polyahl, a processing aid, and water wherein:

- 10 (a) the polyahl, comprising a polyoxyalkylene polyol, has at least two isocyanate-reactive hydrogen atoms/molecule, an average equivalent weight of from 1600 to 5000, an average oxyethylene content of at least 10 percent based on its total weight, and an average primary hydroxyl content of at least 55 percent of its total hydroxyl content;
- 15 b) the processing aid, comprising one or more monohydroxyl polyethers having an equivalent weight of from 600 to 5000 and an oxyethylene content of from 25 to 75 weight percent, is present in an amount of from 1 to 20 parts per 100 parts by weight of (a);
- c) the water is present in an amount of from 2 to 10 parts per 100 parts by weight of (a).

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INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 95/10717

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08G18/28 //(C08G18/28,101:00)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	FR,A,2 268 036 (NAPHTACHIMIE) 14 November 1975 see claims 1,3 see page 2, line 33 - page 3, line 9 see page 4, line 5 - line 26 ---	1,2,8,9
X	WO,A,94 04586 (DOW) 3 March 1994 see claims 1-6,8-12 see page 5, line 14 - page 7, line 27 see page 9, line 15 - line 20 ---	1,2,6,8,9
A	FR,A,1 487 458 (NAPHTACHIMIE) 7 July 1967 see page 3, right column, paragraph 1 -paragraph 5 see page 2, left column, paragraph 4 -paragraph 7 -----	1

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
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- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
- *&* document member of the same patent family

Date of the actual completion of the international search

14 December 1995

Date of mailing of the international search report

29.12.95

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl,
Fax (+ 31-70) 340-3016

Authorized officer

Van Puymbroeck, M

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 95/10717

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
FR-A-2268036	14-11-75	AT-B- 344403 BE-A- 828095 CH-A- 602816 DE-A- 2517281 GB-A- 1480972 NL-A- 7504635 SE-A- 7503966	25-07-78 20-10-75 15-08-78 23-10-75 27-07-77 21-10-75 20-10-75
WO-A-9404586	03-03-94	NONE	
FR-A-1487458	26-10-67	NONE	